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21 July 1965 - 28 February 1966

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For

MANNED SPACECRAFT CENTER  
HOUSTON, TEXAS

NASA Manned Spacecraft Center  
Power Generation Branch  
2101 Webster-Seabrook Road  
Houston, Texas 77058



MIDWEST RESEARCH INSTITUTE

425 VOLKER BOULEVARD/KANSAS CITY, MISSOURI 64110/AC 816 LO 1-0202

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by

T. D. Bath  
A. D. McElroy

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## PREFACE

Personnel contributing to this program are I. C. Smith, T. D. Bath and Project Leader, A. D. McElroy. Dr. Jerome Brewer of the Institute, Dr. W. H. Schechter, and Dr. Frank Fowler served as consultants in various phases of the program.

Approved for:

MIDWEST RESEARCH INSTITUTE



F. V. Morriss, Director  
Chemistry Division

23 February 1966

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## SUMMARY

Purities of hydrogen and oxygen delivered to the Apollo fuel cell are a potential source of variability in performance. Means of effecting purification in-flight were evaluated. Several techniques were screened for suitability. Those which appeared to be workable were further studied. Purifications which are judged to be feasible are removal of carbon dioxide and carbon monoxide from oxygen, and separation of hydrogen from impurities by diffusion through metallic membranes. No technique was found to be capable of dealing with inert impurities in oxygen.

The basic units required for removing  $\text{CO}_2$ , CO, and methane from oxygen are a lithium hydroxide absorber for  $\text{CO}_2$ , a humidifier, and a catalytic burner. The function of the catalytic burner is combustion of CO to  $\text{CO}_2$  and of  $\text{CH}_4$  to  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . Carbon dioxide in very dry air or oxygen is not effectively absorbed by lithium hydroxide, and a humidifier is thus required. The three units would be placed in line, in the order: burner, humidifier and absorber. For the Apollo mission the combined weights of these units and accessory equipment would be about 45 lb. Parasitic power consumption is estimated to be 10 KWH, or 1.7 per cent of design power output. If absorption of  $\text{CO}_2$  is the only desired purification, the package would weigh about 17 lb., and total parasitic power consumption would be about 5.4 KWH. A third possible package contains a burner designed to oxidize only carbon monoxide; the weight and power requirements of this package are estimated to be 30 lb. and 6 KWH.

Purification of 99.5 per cent hydrogen by diffusion through metallic membranes results, according to our analysis, in a delivered power of 576 KWH rather than the mission goal of 600 KWH. The delivered power is substantially higher than that possible with a purge schedule revised for 99.5 per cent hydrogen. The purification technique thus offers a partial solution to a possible purity problem. The advantages of purification are minimized further by 105 lb. of added weight, and by added operational complexity. For missions where design freedom exists, one concludes that impurities in hydrogen can be better handled by increasing the initial quantity of hydrogen and by revising purge schedules.

Data specifically relevant to operating conditions inherent in the Apollo mission were not usually available, and developmental studies of worthwhile purification packages will be required. Estimates of the time and manpower required for development are included in the report.

## I. INTRODUCTION

The hydrogen-oxygen fuel cell is presently an essential part of the electrical power system in space vehicles having missions of longer duration than one or two days. Fuel cells for use in space missions must be very reliable and steady performers, and should be able to obtain a maximum of power per unit weight of fuel. Power efficiency can be affected by the quality or purity of the fuels. Power efficiency can be reduced either by the necessity to vent fuel along with useless or inert impurities, or by cell malfunction due to undesired chemical or physical phenomena traceable to impurities.

It is self-evident that purity is at a premium in space missions, and that rigorous control of purity will be necessary from manufacturing through storage to actual use in the fuel cell. In the current space program every effort is being made to insure adequate control of purity. This program was undertaken in an effort to supplement the normal techniques required to maintain purity, and in part to assess the feasibility and desirability of removing impurities during rather than before a mission in space. In brief, the current program has the objective of evaluating possible techniques of effecting various purifications between fuel storage systems and fuel cells. The program is primarily concerned with the Apollo mission. Studies therefore have been subject to certain restrictions that may not be applicable to future, less completely designed missions. Generally, however, the results are applicable to all missions specifically requiring hydrogen-oxygen fuel cells.

The scope of the program is as follows. Hydrogen with a minimum purity of 99.5 per cent and oxygen with a minimum purity of 99.6 per cent were assumed to be available as fuels for the Apollo fuel cell. Various impurities were specified in each fuel; these consisted chiefly of carbon monoxide, carbon dioxide, methane and inert gases in oxygen, and of inert gases including methane in hydrogen. The program consisted of paper evaluation of the feasibility of various methods for removing any or all of the impurities. Included in the evaluation are suggested designs, with appropriate design and operating parameters, estimates of the expense and time required for finalization of feasible systems, and estimates of feasibility in terms of uncertainties in design or operating characteristics. Suggested devices must operate without disturbance of the fuel cell proper, with the exception that electrical power is available as needed.

## II. DISCUSSION

### II-A. General Considerations

II-A-1. Purities: Specified purities for oxygen are listed in Table I as per MSFC-SPEC-399. The minimum supply purity is 99.60 per cent, with impurities as specified for Grade B breathing oxygen. The purification device(s) should strive for a delivered purity of 99.995 per cent as per Grade A fuel cell oxygen. The supply impurities can thus be as high as 4,000 ppm, and impurities as delivered to the fuel cell ideally no more than 50 ppm.

TABLE I

#### OXYGEN REQUIREMENTS

<u>Requirement</u>	<u>Grade A for Fuel Cells</u>	<u>Grade B for Breathing</u>
Purity	99.995	99.60
Methane and ethane	Methane 10 ppm; Ethane 2.0 ppm	Methane 25.0 ppm; Ethane 2.0 ppm
Propane and higher hydrocarbons	1 ppm as propane	1.0 ppm as propane
Alkyne and acetylene hydrocarbons	0.05 ppm as acetylene	0.05 ppm as acetylene
Total hydrocarbons	14.0 ppm as methane	29.0 ppm as methane
Moisture	3.0 ppm	3.0 ppm
Nitrous oxide	1.0 ppm	1.0 ppm
Halogenated hydrocarbons	1.0 ppm	1.0 ppm
Odor	No odor	No odor
CO and CO <sub>2</sub>	1.0 ppm total	5.0 ppm carbon monoxide; 5.0 ppm carbon dioxide

Notes: 1. For Grade A oxygen - all other impurities including inert gases and nitrogen, 30 ppm.

2. For Grade B oxygen - other impurities, 3,956 ppm by difference.



The impurities to be removed, as obtained by difference from values given in Table I, are methane, carbon monoxide, carbon dioxide, and inert gases. The amounts of each to be removed are listed in Table II.

TABLE II

DESIRED PURIFICATION OF OXYGEN, IN PPM

	<u>Supply</u> <u>Oxygen</u>	<u>Delivered</u> <u>Oxygen</u>	<u>Difference</u>
CH <sub>4</sub>	25	10	15
CO	5 }	1 combined	9 combined
CO <sub>2</sub>	5 }		
Inerts	3,956	30	3,926

Of these impurities, carbon monoxide and carbon dioxide are of greatest concern. Carbon dioxide causes cell fouling and malfunction through formation of insoluble carbonates. Carbon monoxide is expected to oxidize to carbon dioxide at or near the electrode surfaces, and thus should be as objectionable as carbon dioxide. Methane is nominally an inert, though it is subject to catalytic oxidation at temperatures not much higher than the cell operating temperature of 450°F. The inert gases and methane are undesirable in that they must be vented, along with some oxygen, in order to maintain the partial pressure of oxygen at the electrodes at an acceptable value.

The permissible impurities in fuel cell grade hydrogen are shown in Table III. The impurities to be removed total 4,950 ppm. The quantity of each impurity to be eliminated has not been specified; they consist of water and inerts, including methane.

TABLE III

PERMISSIBLE IMPURITIES IN HYDROGEN

Total gaseous impurities	50 ppm by volume
Selected impurities, including nitrogen, water and methane	9.0 ppm by volume
Specific impurities:	
Oxygen plus argon, maximum	1.0 ppm by volume
Helium, maximum	39.0 ppm by volume
Carbon bearing gases (CO and CO <sub>2</sub> )	1.0 ppm by volume

II-A-2. Other constraints: Additional constraints involve operating parameters of the fuel cell, fuel storage systems, and environmental conditions within the control module. One specific constraint of considerable importance (for the Apollo mission) is that the fuel cell proper together with its accessories cannot be modified (i.e., the heat exchanger of the fuel cell cannot be tapped into, and hydrogen or oxygen cannot be passed through the fuel cell preheaters before introduction to purifiers). Other operating constraints, either as specified in the contract or as obtained later from the sponsor, are listed in Table IV. A complete list of constraints might include all the operating parameters of the fuel cell, fuel storage system, and related vehicular parts. A minority of these are limiting for our purposes, and these are presented in Table IV.

TABLE IV  
OPERATIONAL CONSTRAINTS

	<u>Oxygen</u>	<u>Hydrogen</u>
Storage temperature, °F	-300 to +150	-425 to +150
Pressure delivered to pressure regulator, psia	150-1020	100-295
Minimum acceptable pressure delivered to pressure regulator <sup>a/</sup>	100	100
Flow rates, lb/hr		
Minimum	1.2	0.15
Maximum	3.33	0.432
Average	1.32	0.162
Lb/KWH	0.685	0.0856
Lb vented/KWH at 99.99 per cent purity	0.00346	0.00432
Maximum temperature of inlet pressure regulator, °F	180	180

<sup>a/</sup> Certain purification devices might require in-line reduction of pressure.  
A value of 100 psia was selected as a permissible lower limit.

Other data of direct or indirect significance are summarized below:

Fuel cell operating temperature	450°F
Fuel cell operating pressure	50 psia
Mission duration	14 days/336 hr.
Fuel consumption	
Oxygen	411 lb.
Hydrogen	51.5 lb.
Oxygen supply	640 lb., including breathing O <sub>2</sub>
Hydrogen supply	56 lb.
Fuel cell power	
Overload	6.885 kw
Peak	4.26 kw
Average	1.77 kw
Minimum	1.69 kw
Total power output	600 KWH
Available electrical power	29 v. DC or 115 v., 3Ø 400 cps
Environmental temperature	30° to 160°F

II-A-3. Criteria for assessing feasibility: These are several, and no simple means are available to reduce all criteria to a simple formula. Pertinent factors are given below, with discussion as appropriate.

a. Added weight must be minimized. A permissible weight must, however, be determined in terms of the effectiveness of a device, and the urgency of the need for the device.

b. Volume (occupied space) should also be minimized.

c. Purification devices must operate automatically, with perhaps an occasional manual assist.

d. Performance reliability should be absolute.

e. Purification devices must not upset the systems of checks and balances extant in the power package. They cannot, for example, supply heat energy in quantity sufficient to overload the heat rejection capacity of the fuel cell heat exchange system.

f. Power consumption should be minimum. An acceptable level of power consumption is a function of need. In addition, it is possible that parasitic power expenditure will result in total power outputs equal to or greater than that possible without fuel purification.

g. In addition to a conservative use of power, purification procedures must not be wasteful of fuel.

#### II-A-4. Proposed purification methods:

a. Oxygen: Impurities in oxygen are of two types. Carbon monoxide and carbon dioxide are polar or reactive molecules susceptible to chemical change and to adsorption or absorption. Methane may be classified either as an inert or as an impurity susceptible to oxidation. Remaining impurities are nonpolar and chemically inert, and separative techniques must be purely physical in nature (adsorption, diffusion, etc.) unless one chemically condenses and regenerates oxygen. Methods considered for oxygen purification are as follows:

Absorption of carbon dioxide by conversion to carbonates.

Adsorption of carbon dioxide, e.g., on molecular sieves.

Catalytic combustion of carbon monoxide to carbon dioxide followed by adsorption or absorption.

Catalytic combustion of methane followed by removal of carbon dioxide.

Adsorption of inert gases, including methane.

Chemical removal of oxygen followed by regeneration as molecular oxygen.

b. Hydrogen: The impurities under consideration are water, methane, and inert gases. Methane in hydrogen must be classified as an inert, since methane chemically is a nonpolar molecule subject only to oxidation, or to pyrolytic decomposition at unacceptably high temperatures. Water is polar and reactive, and thus could be removed by adsorption or reaction. Since the hydrogen stream becomes rich in water in the fuel cell system, we have not seriously considered its removal.

The methane and inert gases constitute a problem from the standpoint of power efficiency, and a device capable of removing them could result in increased power efficiency. Approaches considered for removal of these impurities are as follows.

Diffusion through semi-permeable membranes reportedly yields hydrogen very high in purity. Filtration of liquid hydrogen should separate impurities as frozen solids. Distillation (selective condensation) at temperatures below the critical point should remove all but traces of impurities. Selective adsorption on adsorbents such as silica gel is an effective means of purifying hydrogen. Metal hydride formation and decomposition is a chemical means of separating hydrogen from impurities.

## II-B. Preliminary Evaluations

The techniques listed in II-A-4 were rough-analyzed to assess feasibility in terms of various constraints. This analysis eliminated all but a few from further serious consideration.

II-B-1. Adsorptive techniques: Adsorption differs from absorption in that the former is chiefly physical in nature, while absorption involves definite compound formation with the absorbate. While both processes involve equilibria, absorption processes usually have equilibrium values permissive of more complete removal of substances than do adsorption processes.

Adsorption is most effective for polar molecules. Water would, therefore, be the most effectively adsorbed of all the impurities in hydrogen or oxygen. Carbon dioxide is less effectively adsorbed than water, and carbon monoxide less effectively than carbon dioxide. Water may, in fact, be very effectively adsorbed at ambient temperatures. The Linde molecular sieves have high capacities for water, and the capacities are sufficient at low partial pressures of water that removal of trace quantities of water can be accomplished with reasonable quantities of molecular sieve. In Linde Form 9690-C, Data Sheet No. 5A-3, dew points of  $-100$  to  $-120^{\circ}\text{F}$  are indicated to be obtainable with loadings of 3 - 4 per cent by weight of water. These dew points are equivalent to about 1 ppm in air at 1 atmosphere. In oxygen at 50 atmospheres, the water concentration would be about 1/50 ppm. At 100 ppm water by volume (an arbitrarily assumed value), the total oxygen fed to the Apollo fuel cell would contain about 0.025 lb. of water. Approximately 1 lb. of 5A Molecular Sieve would be theoretically sufficient to reduce the water content for the entire mission to substantially less than 1 ppm, with adsorption occurring at temperatures listed as ambient in the Apollo module. We have not further refined these data in a device for removing water. At this point, we believe it sufficient to point out that a water removal device could be quickly and simply designed, if the need is real.

The available data for carbon dioxide adsorption do not contain information at low pressures of carbon dioxide. For our purposes, a partial pressure of about 0.05 mm.  $\text{CO}_2$  in oxygen at 50 atmospheres would be required.

Extrapolation of available data (Linde Form 9691-E, Data Sheet No. 24) to low pressures indicates that CO<sub>2</sub> might adsorb on 5A Molecular Sieve as desired. This uncertainty, together with the facts that (1) water will drive off adsorbed CO<sub>2</sub>, and (2) that absorption of CO<sub>2</sub> on lithium hydroxide has been amply demonstrated, has led us to the conclusion that adsorption of CO<sub>2</sub> should not be seriously considered.

For inert impurities in oxygen, adsorption is almost the only applicable technique, and adsorption is also applicable in theory to inerts in hydrogen. Two serious drawbacks exist, namely, that we are limited to ambient conditions where inerts absorb very poorly, and that a system of recycle streams is required if the major constituent (O<sub>2</sub> or H<sub>2</sub>) is to be recovered without waste. At essentially cryogenic temperatures inert impurities can be very effectively removed in earth-bound plants in which weight and complexity are of minor importance, and yield is considerably less important than effectiveness.

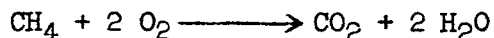
For example, pure methane at 200 mm. pressure absorbs to the extent of 0.0001 per cent by weight on 4A Molecular Sieve at 77°F (Linde Form 9692-C, Data Sheet No. 71). The adsorption of methane is thus not possible at ambient conditions. At low temperatures, methane adsorbs very effectively on silica gel, synthetic zeolites and charcoal, as indicated by data reported by Hiza and Kidnay.<sup>3/</sup> At about 0.1 mm. and -310°F, methane adsorbs on these substrates to the extent of 3 - 8 per cent. Data pertinent to the low temperature adsorption characteristics of nitrogen and methane in hydrogen have also been obtained by Hiza and Kidnay.<sup>4/</sup> Briefly, the data reveal an effective adsorption at low temperatures of nitrogen and methane in hydrogen. This capacity disappears with elevation of temperature, however.

One concludes, therefore, that adsorptive techniques are suitable for water and possibly for carbon dioxide, but are completely unsuitable for inert (nonpolar, nonreactive) contaminants.

II-B-2. Absorption of carbon dioxide: Absorption of carbon dioxide by lithium hydroxide was selected as the preferred method of removing this impurity from oxygen. Lithium hydroxide is available in a form suitable for canister use, and its effectiveness and reliability have been proven.<sup>23/</sup> Since carbon dioxide is a very objectionable impurity, it is essential that an absorbent be theoretically and practically able to reduce CO<sub>2</sub> to a very low level. Of equal importance in the absorbing system is freedom of interfering or competitive reactions. The lithium hydroxide method is both absolute and devoid of competitive reactions.

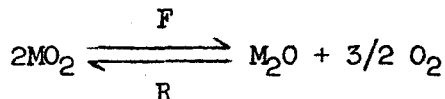
II-B-3. Catalytic oxidation of carbon monoxide: The catalytic oxidation of carbon monoxide can be effected over a wide variety of catalysts,<sup>10/</sup> chiefly oxides of transition metals. Elevated temperatures are not a requirement, though burners now in use employ somewhat elevated temperatures to insure combustion of hydrocarbons, excluding methane.<sup>11/</sup> The operational units employ Hopcalite, a commercially available mixture of copper and manganese oxides. We have selected Hopcalite over other possible catalysts on the basis of its availability and an extensive successful operating history. The catalytic oxidation of carbon monoxide appears to be quite feasible, and detailed analyses have therefore been conducted.

II-B-4. Catalytic oxidation of methane: Methane is considerably more difficult to oxidize than are other hydrocarbons.



There is no question, however, that complete methane oxidation can be effected if sufficiently high temperatures are employed. Since methane can be treated in the basic package used for oxidation of carbon monoxide, we have conducted detailed analyses which are presented in a later section of the report.

II-B-5. Chemical sequestering of oxygen: Certain of the highly electropositive metals react with molecular oxygen to form superoxides,<sup>5,6/</sup> e.g.,  $\text{NaO}_2$ , which dissociate under the influence of heat to oxygen, plus metal oxide or peroxide. The only superoxides sufficiently defined for our purposes are those of sodium and the heavier alkali metals. This chemistry could serve as a basis for purification, although a distinct disadvantage lies in the fact that the major component must be processed. Process chemistry is depicted by the following equation:



The forward reaction (F) would require either a high temperature or a low pressure (or both), relative to the temperature and pressure of the reverse (R) reaction. The inert gases would, according to this scheme, be swept out prior to decomposing the superoxide. Carbon dioxide should form carbonates; whether or not carbon dioxide removal would be permanent depends on the stability of the carbonate under conditions required for the forward (F) reaction. Carbon monoxide and methane are theoretically oxidizable to  $\text{CO}_2$  in this system; we do not know whether they, in fact, would be.

Data necessary for a complete evaluation of this technique are not available. The available data are, however, sufficient to justify the conclusion that detailed studies need not be made. Stephanou et al.<sup>6/</sup> have shown that a temperature of about 750°F is required for conversion of sodium peroxide ( $\text{Na}_2\text{O}_2$ ) to sodium superoxide. Oxygen pressures greater than 200 atmospheres were employed in reactions with gram quantities, which required up to 100 hr. for completion. These facts alone suffice to eliminate sodium oxides from further consideration.

Potassium superoxide is too stable for our purposes, as it can be melted at about 750°F.<sup>7,8/</sup> Dissociation pressures of  $\text{KO}_2$  are of the order of a few millimeters of Hg at 600°F, and less than 1 atmosphere at 840°F.<sup>8/</sup> Dissociation pressures of at least 100 psia would be required in a purifying unit, and this surely means temperatures in excess of 1000°F. We have not, in this and other situations, felt that gas compressors should be employed, since a compressor adds complexity, reduces reliability, and requires a substantial outlay of energy or power. In this particular instance, compression of oxygen from 0.1 atmosphere to 10 atmospheres would require a total expenditure of more than 20 per cent of the electrical power generable by the Apollo fuel cell power plant.

Regenerable oxygen recovery systems are the objective of current research relevant to space missions. In these studies the objective has been recovery of oxygen from breathing air, and the systems necessarily are capable of regenerating oxygen. The  $\text{CoO-Co}_3\text{O}_4\text{-O}_2$  system has been investigated by the Dynatech Corporation.<sup>9/</sup> Oxygen converts  $\text{CoO}$  to  $\text{Co}_3\text{O}_4$ , and reduction of pressure combined with slight temperature elevation results in release of oxygen. The  $\text{CoO-O}_2$  reaction was carried out at 1750°F, and regeneration of oxygen was effected at 1800°F under reduced pressure. These temperature requirements are much too high for our purposes, as energy consumed in heating oxygen would be prohibitively high.

In summary, we conclude that chemical sequestering of oxygen is not a suitable method for purifying fuel cell oxygen.

II-B-6. Purification of hydrogen by diffusion through metallic membranes survived preliminary evaluations, and was considered in further detail. Information obtained from the literature and from manufacturers of these purifiers indicated excellent purification at a nominal expense of energy and probably without undue loss of fuel.

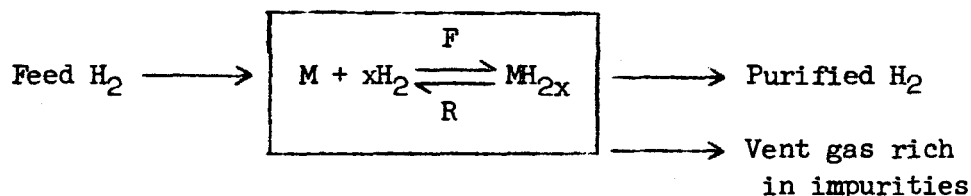
II-B-7. Filtration of liquid hydrogen was eliminated as a purification technique. When this suggestion was advanced it had been assumed that hydrogen would be maintained in the liquid state throughout the mission.



Since hydrogen will be supercritical throughout the mission, filtration is not applicable. This technique might be considered as a part of the fuel loading procedure, as literature data<sup>1,2/</sup> indicate impurities, with the exception of helium, to be both solid and insoluble at liquid hydrogen temperatures. We are of the opinion, however, that filtration would be difficult to engineer. Furthermore, this technique will not eliminate the storage containers as a probable source of impurities.

II-B-8. Distillation: Condensation of hydrogen was stricken from the list for the reasons listed in II-B-2. Again, impurities are quite non-volatile<sup>1,2/</sup> at liquid hydrogen temperatures, and the technique is theoretically sound. Its effectiveness would be limited by the cleanliness of a final storage tank.

II-B-9. Hydride formation: The basic features of this proposed technique are shown below.



Forward reaction: Low temperature and/or high pressure.

Reverse reaction: Low pressure and/or high temperature.

The technique requires an  $\text{M-MH}_x$  combination which reverses readily at reasonable conditions of temperature and pressure. The beryllium-hydrogen ( $\text{BeH}_2$ ) and boron-hydrogen ( $\text{B}_2\text{H}_6$ ) systems would be best from a weight standpoint, but neither beryllium or hydrogen is known to react with hydrogen except possibly at extremes of temperature and pressure. Lithium ( $\text{LiH}$ ) is next in line from a weight standpoint, but must be ruled out on the basis of high temperature required for the dissociative reverse reaction. The dissociation pressure of lithium hydride is 1 atmosphere at ca.  $1550^\circ\text{F}$ , and less than 1 mm. at  $930^\circ\text{F}$ .<sup>12/</sup> Sodium hydride dissociates at lower temperatures; the dissociation pressure of sodium hydride is 1 atmosphere at about  $800^\circ\text{F}$ .<sup>13/</sup> Dissociation pressures are 1 atmosphere at  $804^\circ\text{F}$  for potassium hydride<sup>14/</sup> and at  $831^\circ\text{F}$  for rubidium hydride.<sup>15/</sup> A minimum allowable dissociation pressure is for our purposes 50 psia, the fuel cell operating pressure. Dissociation pressures of 50 psia for the various hydrides are achieved at  $870^\circ\text{F}$  for  $\text{NaH}$ , at  $894^\circ\text{F}$  for  $\text{KH}$  and at  $950^\circ\text{F}$  for  $\text{RbH}$ .

In addition to the requirement of relatively high temperatures for dissociation of the alkali metal hydrides, the alkali metals require hydriding temperatures ranging from a minimum of about 400°F for sodium and potassium to about 1000°F for rubidium. At these temperatures the metals are liquid, and severe handling and corrosion problems can be expected. The alkali metals were as a consequence not seriously considered.

The uranium-hydrogen-UH<sub>3</sub> system offers no advantage over alkali metal systems from a temperature standpoint. However, uranium metal prepared by decomposition of UH<sub>3</sub> is a very finely divided solid which reacts rapidly and completely with hydrogen even at 77°F.<sup>16,17/</sup> The dissociation pressure of UH<sub>3</sub> is 50 psia at about 900°F. The heat of formation of UH<sub>3</sub> is approximately -30 kcal/mole, which means that this much energy must be dissipated during the formative reaction and supplied during the dissociation reaction.

In spite of the fact that the temperature requirement appeared to be too high, we elected to proceed with further evaluations of the uranium-hydrogen-uranium hydride system. The results of this evaluation are discussed in the next section of this report.

### III. ENGINEERING EVALUATIONS OF TENTATIVE PROCESSES

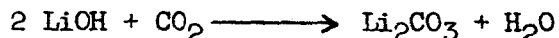
As stated earlier, oxygen of 99.6 per cent purity and hydrogen of 99.5 per cent purity were assumed as the initial basis for the evaluation. The concentrations of the various contaminants are discussed earlier in the report. One would like to be able to simply compare the fuel, weight, space, and power requirements for each system and decide which are most attractive simply on this basis. Although this can be done to a certain degree, intangible factors, such as the reliability of the fuel cell system, must be considered in proper proportion with the material factors.

There are five systems which have been selected for further evaluations, two pertaining to hydrogen purification and three concerned with removing certain impurities from oxygen. The material requirements of these systems are described and compared with the requirements for operating the fuel cell without any purification devices. The systems which will be considered are: (A) Oxygen purification: (1) carbon dioxide removal, (2) low temperature oxidation, and (3) high temperature oxidation. (B) Hydrogen purification: (1) the uranium hydride process, and (2) diffusion through metallic membranes. The following section of this report describes the designs which have been developed for these processes. The designs must be viewed as tentative, since design data at mission conditions were not always available. The relative merits of the various systems will be evaluated in the third section, and additional features of the design of various devices are presented in the Appendix.

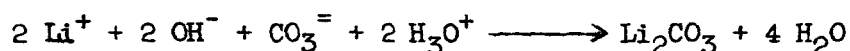
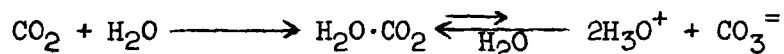
### III-A. Design of Purification Systems

III-A-1. Purification of oxygen: The impurities which may appear in the oxygen stream may be divided into three categories: inerts, incompletely oxidized carbon compounds, and carbon dioxide. The most undesirable impurity is carbon dioxide since it can react directly with the electrolyte of the fuel cell to cause fouling or malfunction of the system. Of the compounds of the second category carbon monoxide is the most objectionable since it will almost certainly oxidize to the dioxide in the fuel cell, giving rise to the formation of insoluble carbonates. The other compounds, mainly hydrocarbons, become harder and harder to oxidize as molecular weight decreases, with methane presenting the least hazard to cell operation. No technique has been developed which will make it possible to remove inerts from the oxygen stream without unreasonable expenditure of energy, mass, and fuel. We are thus concerned only with removal of carbon dioxide and the other carbon compounds which may be oxidized to carbon dioxide. The purification schemes are of three types: (1) absorption of initial carbon dioxide, (2) low temperature oxidation followed by absorption, and (3) high temperature oxidation followed by absorption. In the first scheme only the original CO<sub>2</sub> in the stream is removed. The second operation will provide enough oxidation to completely oxidize all of the CO to CO<sub>2</sub> which will then be removed by absorption. There will be some oxidation of hydrocarbons at this level. The final type of operation is designed to oxidize 60 per cent of the methane present to CO<sub>2</sub>. It should completely oxidize all other hydrocarbons. The CO<sub>2</sub> formed will then be removed by absorption.

a. Simple removal of carbon dioxide: This process uses a packed bed of lithium hydroxide granules to remove carbon dioxide from the gas stream by formation of the carbonate.



The reaction is carried out at ambient temperature (50°F). At this temperature the reaction goes completely to the right, since the decomposition pressure of Li<sub>2</sub>CO<sub>3</sub> is completely negligible. In order to absorb the carbon dioxide it is important that there be a certain amount of moisture in the gas. This has been substantiated by a number of sources.<sup>18-20/</sup> The role of water is not fully understood. It is reasonable to postulate that water and carbon dioxide co-adsorb, and that carbonic acid is thus the species which reacts with lithium hydroxide.



Available information indicates that the effectiveness of the reaction increases linearly with the relative humidity of the system up to approximately 10 per cent R.H., after which water has no further effect. The present design is based on maintaining at least 8 per cent R.H. in the system by means of desorbing water from a column of silica gel.

There are some areas of uncertainty in extrapolating available information to a design under these conditions. Most important are the effects of high oxygen partial pressure and high system total pressure. No data are available on the effects of either of these variables on the system kinetics or the amount of water vapor necessary to make the reaction most effective. In order to reach final decisions on the design of this system it would be necessary to experimentally evaluate the system under the conditions of interest. A schematic flow diagram of the system is shown in Fig. 1. The feed temperature was assumed to be  $-135^{\circ}\text{F}$  or greater and the system pressure 800 psia or less. The temperature was arrived at by assuming the same driving force for heat transfer as brought about the increase in hydrogen feed temperature between the storage tank and the preheater. The size of the humidifier was based on an operating pressure of 150 psia. The material requirements of the system are:

Weight	- 17 lb.
Displacement	- 1 ft. <sup>3</sup>
Average Power	- 16 w.
Maximum Power	- 40 w.

This design is a conservative estimate based on available data. Experimental data would be important in any further attempts to refine the design.

b. Low temperature oxidation: The low temperature oxidation system is designed to convert essentially all of the CO in the gas stream to CO<sub>2</sub> so that it can be removed by absorption. The oxidation is accomplished over Hopcalite catalyst at  $70^{\circ}\text{F}$ . The kinetic data used were taken from a dissertation by A. G. Mulay.<sup>10/</sup> A schematic flow diagram of the system is shown in Fig. 2. The slight increase in gas stream temperature and CO<sub>2</sub> content does not materially affect either the humidifier or the absorber. Thus, we find the material requirements to be:

Weight	- 30 lb.
Displacement	- 1.5 ft. <sup>3</sup>
Average Power	- 18 w.
Maximum Power	- 45 w.

The increase in weight is due principally to the addition of the reactor for the oxidation. In many ways this system seems quite attractive. However, here again, it should be stressed that no data exist on precisely this system and experimental verification of the design is necessary.

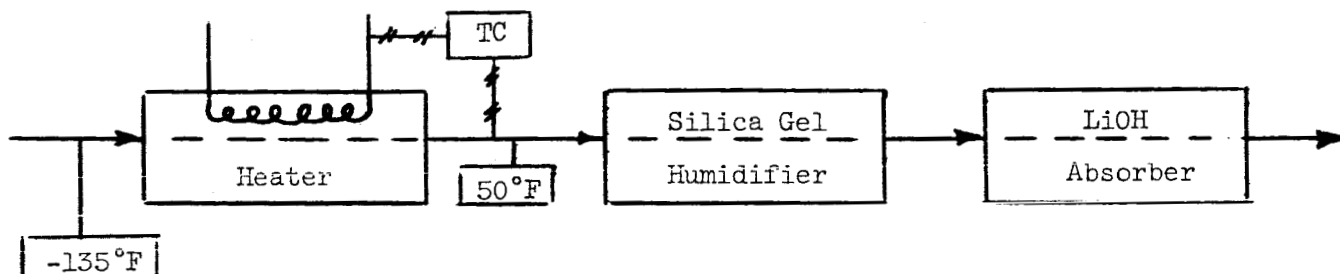


Fig. 1 - Carbon Dioxide Absorber

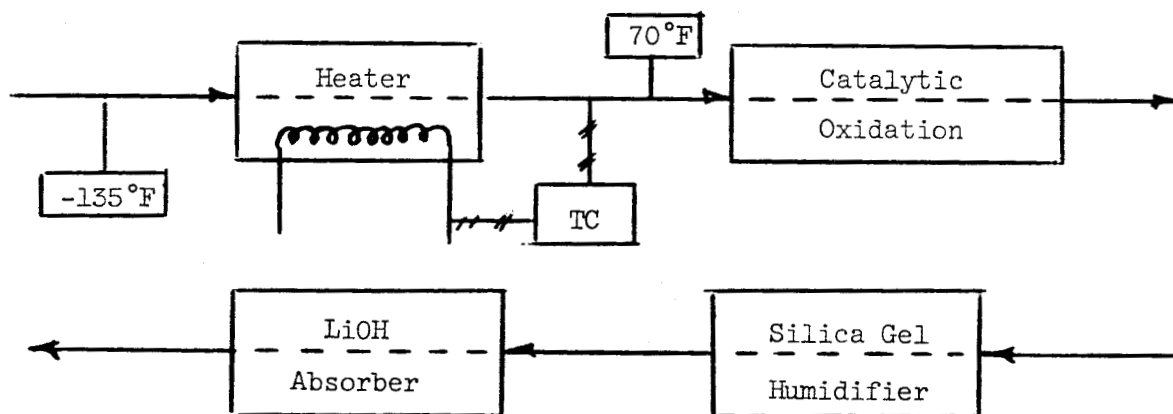


Fig. 2 - CO Burner and  $\text{CO}_2$  Scrubber

c. High temperature oxidation: The basic criterion for the design of the high temperature system is that the temperature in the Hopcalite reactor be high enough that 60 per cent of the methane in the system be converted to  $\text{CO}_2$ . There is some discrepancy in the literature regarding what this temperature is. There is no doubt that it is a moderately high temperature, but Johns<sup>21/</sup> suggests 550°F while MSA data<sup>22/</sup> suggest a temperature of 700°F. In order that the design be conservative it was decided to use the 700°F operating temperature as a basis. Temperatures this high require that heat losses from the system to the surroundings be taken into account. Another important addition here is the use of a countercurrent heat exchanger to remove heat from the reactor effluent and pass it into the reactor feed. A schematic flow diagram of the high temperature oxidation system is shown in Fig. 3.

The material requirements of the system are as follows:

Weight - 45 lb.  
 Displacement - 2 ft.<sup>3</sup>  
 Average Power - 30 w.  
 Maximum Power - 60 w.

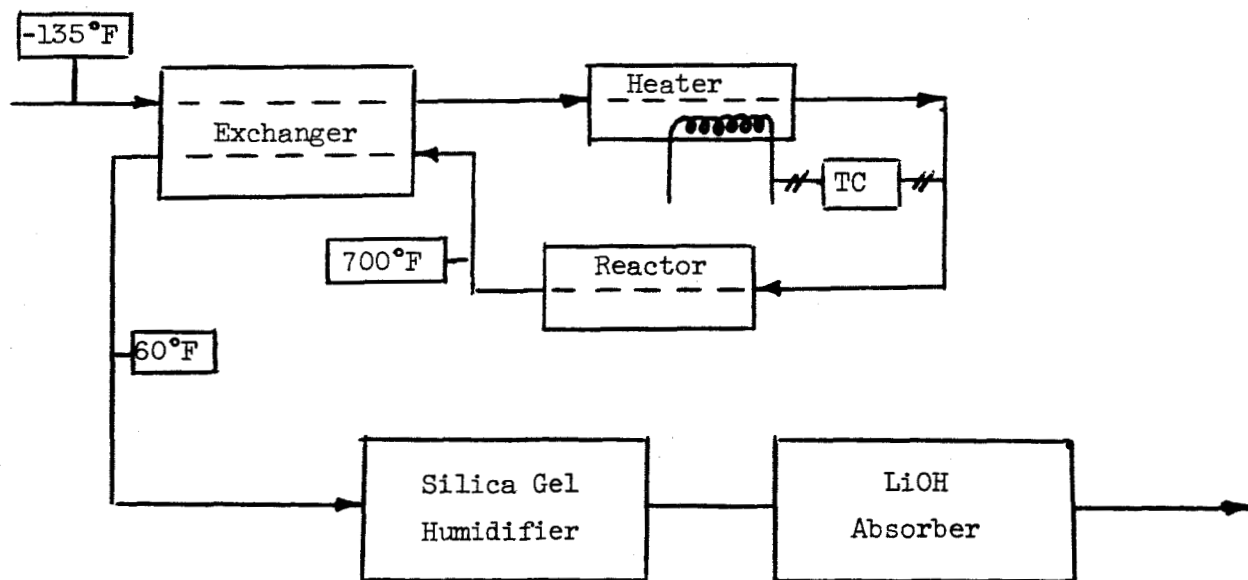


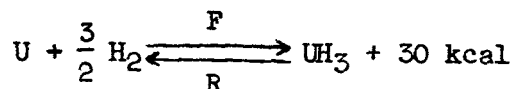
Fig. 3 - Burner for Partial Combustion of Methane

Much of the mass requirement is from the heat exchanger. The mass of catalyst in the reactor is only 3 lb. This system gives maximum assurance that no CO<sub>2</sub> will be formed in the fuel cell, since it should remove all potential sources of CO<sub>2</sub> except 10 ppm of methane.

Each of the three systems for purifying oxygen discussed above is a flow through system, so that there is no loss of oxygen corresponding to the fuel loss in the hydrogen purification systems. Unfortunately, however, these purification schemes do not remove inert impurities from the oxygen so that a venting requirement remains.

### III-A-2. Purification of hydrogen

a. Uranium hydride process: One of the means by which hydrogen may be purified is to withdraw the hydrogen selectively from the impure gas stream by means of a chemical reaction which is specific for hydrogen. When the reaction is reversed, hydrogen is liberated.



This purification system would take advantage of the pressure drop between the hydrogen storage cylinder and the fuel cell. The forward reaction would take place at about 250 psia and 1300°F, while the reverse reaction would be run at about 100 psia and 1100°F. The reaction would be carried out in a two-chamber vessel, with uranium hydride being alternately formed and then decomposed. Two chambers are required for continuous generation of hydrogen. Conductive fins extending into both vessels would provide heat transfer surfaces for heat flux from the reacting chamber to the chamber in which UH<sub>3</sub> is being decomposed. Some additional heat will have to be added to the system. This will be minimized by the use of an exchanger between the impure gas entering the reactor and the pure gas leaving the reactor.

A schematic process flow diagram of the system is shown in Fig. 4. The two reactors would operate on a 10-min. cycle, forming UH<sub>3</sub> for 5 min. and then reversing the reaction for 5 min. The design is based on 30 per cent conversion of UH<sub>3</sub> to U at maximum flow rates, which is the highest value consistent with conservative design. Based on this assumption and available rate data the total uranium required is about 12 lb. Due to the high operating temperature of the reactor, the economizing heat exchanger is called on to transfer a great deal of heat which in turn requires considerable transfer area. In addition to this, we have a fairly large heater and several

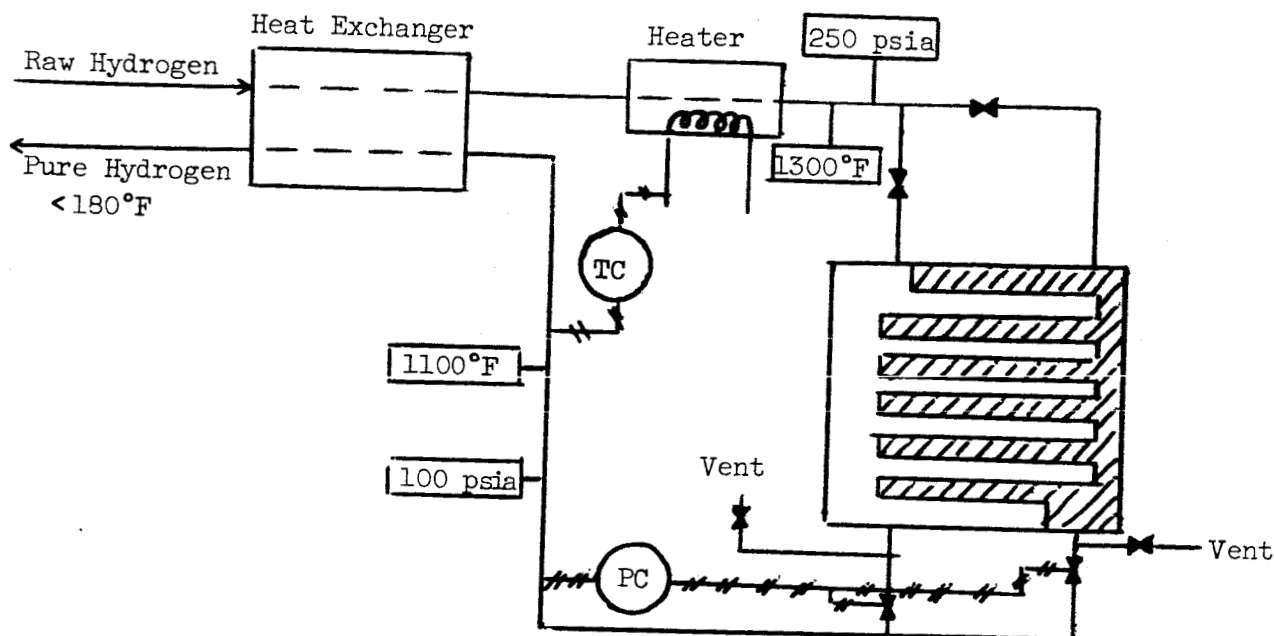


Fig. 4 - Uranium Hydride Purification System

valves and controlling instruments. The total estimated mass of these objects is approximately 190 lb. The energy requirement is 280 w. maximum and 127 w. on the average. Because the process involves the cyclic operation of a chemical reaction, there could be considerable difficulty in control of the product quality. Development of a completely reliable system would require considerable experimental effort.

The degree of purification resulting from the hydride process is limited by the degree to which the unreacted impurities can be removed from the reactor. Calculations indicate that since it is not practical from an energy standpoint to cool and evacuate the chamber, the best way to remove the impurities is by flushing the gas phase in the reactor with impure feed hydrogen. This will result in a purification factor of 25 (from 0.995 to 0.998) with an attendant 10 per cent loss of hydrogen due to flushing. Thus, the fuel loss would be some 6.1 lb. of hydrogen.

In summary, the process has two subjective difficulties: possible difficulty in control of product quality and the production of only 99.8 per cent pure hydrogen rather than the 99.99 per cent desired. The physical requirements are:



Weight - 190 lb.  
Displacement - 7 ft<sup>3</sup>  
Average Power - 127 w.  
Maximum Power - 280 w.  
Loss of Fuel - 6.1 lb. during mission

b. Membrane diffusion process: The use of palladium-silver alloys as a diffuser affords a practical means of obtaining high purity hydrogen. Hydrogen which passes through the metal diffuser is believed to contain less than 1 part/billion impurities. Until recently the use of diffusers in purifying hydrogen was limited to small scale laboratory preparations. Much higher capacity diffusers have been developed through the use of alloys as the diffuser materials. Diffusers having capacities of 200 scfh are now readily available.

A diffuser having a capacity of 50 scfh has adequate capacity for the problem being considered here. A commercially available diffuser\* having this capacity consists of 84 1/16-in. O.D. palladium alloy tubes contained in a 1.75 in. O.D. stainless steel tube which is 18 in. long. The hydrogen is fed into the stainless steel tube and allowed to diffuse through the wall of the 1/16-in. O.D. palladium alloy tubes. Figure 5 presents a schematic flow diagram of the process. Note the relative simplicity of this process compared to the uranium hydride process. One of the operating variables of the process is the amount of venting which is used (i.e., the impurity concentration allowed on the high pressure side of the diffuser). The optimum maximum impurity concentration was found to be 10 per cent for this particular system. This in turn gives rise to a 4.7 per cent loss of H<sub>2</sub> due to venting. Subjectively, the system should be much easier to operate and more reliable than the UH<sub>3</sub> system discussed previously. In addition, the hydrogen produced is of such quality that no venting of the hydrogen side of the fuel cell should be necessary. The physical requirements are:

Weight - 105 lb.  
Displacement - 3 ft<sup>3</sup>  
Average Power - 108 w.  
Maximum Power - 254 w.  
Loss of Fuel - 2.6 lb. during mission

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\* The Model C-50D diffuser manufactured by the Milton Roy Company, St. Petersburg, Florida, was used as a model. Operating characteristics needed for design were kindly supplied by the Milton Roy Company. It should be understood that other companies have capabilities in this area, and our use of the Milton Roy data does not constitute an endorsement of this company's products to the exclusion of those of other companies.

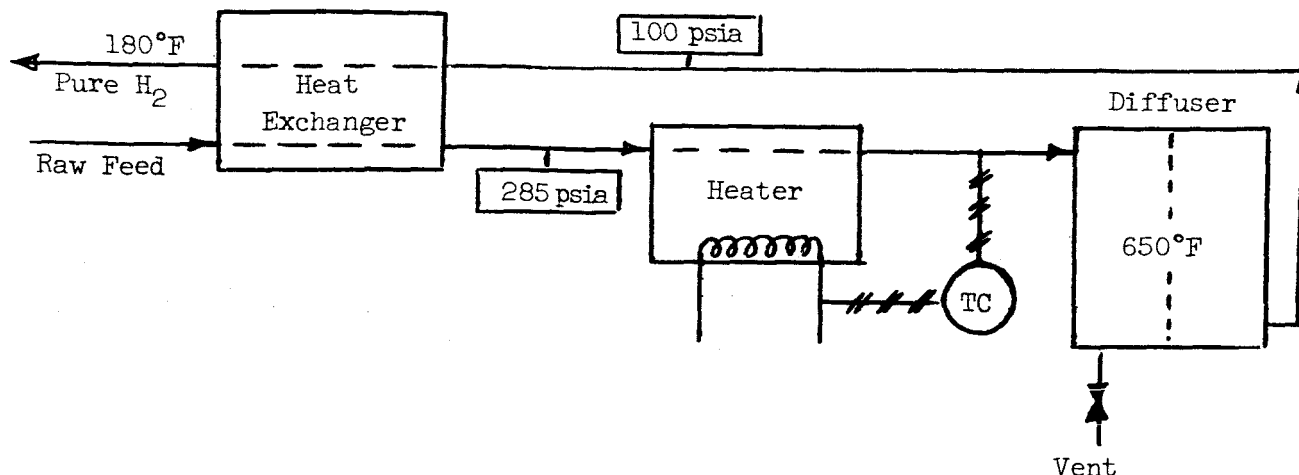


Fig. 5 - Membrane Diffusion Purification Process

### III-B. Comparison of Alternative Purification Schemes

It is difficult to establish a basis for comparison of the schemes for purification of oxygen and hydrogen for two reasons: first, because of the intangible factors mentioned previously; and second, because it is difficult to determine how the fuel cell would operate without the purification devices. Let us first address ourselves to the latter problem. The purge requirements furnished to this program are for gases of 99.99 per cent purity. The question which must be resolved is, "How would the purge system of the fuel cell be operated if the oxygen purity is 99.6 per cent and the hydrogen purity is 99.5 per cent?" In order to have a consistent basis for comparison venting has been calculated on the following basis:

1. The impurity concentration inside the fuel cell was assumed constant at 2.5 per cent. Data furnished by the sponsor (fuel cell purge program report) indicate that in cells flushed on an intermittent basis the impurity concentration ranges from 0 - 5 per cent in a linear fashion.
2. The cell was assumed to be purged continuously at this concentration and at a flow rate which removes impurity as fast as it is introduced.
3. The loss of fuel is calculated from the flow rate determined in (2), assuming the system behaves like a stirred pot.

Having defined this base, we can then enumerate the requirements and advantages of the various processes, compare them to operation without purification and then draw conclusions as to which processes would be preferred.

III-B-1. Oxygen purification processes: The comparison of the several oxygen purification processes is not nearly as straightforward due to the various subjective factors involved. Without purification the direct purge would amount to a total of 84.6 lb. of oxygen for the mission (assuming 99.6 per cent oxygen). However, this purge value will remain constant for all cases. The material requirements for the various schemes are listed in Table V and Table VI.

TABLE V

OPERATING COMPARISONS, FOR 99.6% OXYGEN  
IN THE APOLLO MISSION

	<u>CO<sub>2</sub></u> <u>Absorption</u>	<u>CO</u> <u>Oxidation</u>	<u>CH<sub>4</sub></u> <u>Oxidation</u>	<u>Direct Purge</u>
Weight, lb.	17	30	45	-
Displacement, ft. <sup>3</sup>	1	1.5	2	-
Average Power, w.	16	18	30	-
Maximum Power, w.	40	45	60	-
Loss of Fuel, lb.	84.6	84.6	84.6	84.6
Net Power Available, KWH	477	476	472	482

TABLE VI

OPERATING COMPARISONS, FOR 99.99% OXYGEN  
IN THE APOLLO MISSION

	<u>CO<sub>2</sub></u> <u>Absorption</u>	<u>CO</u> <u>Oxidation</u>	<u>CH<sub>4</sub></u> <u>Oxidation</u>	<u>Direct Purge</u>
Weight, lb.	17	30	45	-
Displacement, ft. <sup>3</sup>	1	1.5	2	-
Average Power, w.	16	18	30	-
Maximum Power, w.	40	45	60	-
Loss of Fuel, lb.	2.6	2.6	2.6	2.6
Net Power Available, KWH	590	589	585	595

III-B-2. Hydrogen purification processes: Without the use of any purification, the purge requirement for the conditions just described amounts to 0.0405 lb/hr or 13.6 lb. of hydrogen for the Apollo mission. This compares unfavorably with 6.1 lb. lost using the uranium hydride process and 2.6 lb. lost in the membrane diffusion process. However, both of these processes require such large amounts of other materials that a simple direct purge program seems attractive unless storage of the additional hydrogen is impractical or impossible.

The three processes are compared in Table VII.

TABLE VII

OPERATING COMPARISONS, FOR 99.5% HYDROGEN  
IN THE APOLLO MISSION

	<u>Uranium Hydride</u>	<u>Membrane Diffusion</u>	<u>Direct Purge</u>
Weight, lb.	190	105	-
Displacement, ft. <sup>3</sup>	7	3	-
Average Power, w.	127	108	-
Maximum Power, w.	280	254	-
Loss of Fuel, lb.	6.1	2.6	13.6
Net Power Available, KWH <sup>a/</sup>	532	576	484

a/ Assuming 55 lb. H<sub>2</sub> available, at 0.0856 lb. H<sub>2</sub>/KWH.

IV. CONCLUSIONS AND RECOMMENDATIONS

We have reached the following conclusions on the basis of analyses of various possible means of purifying fuel cell gases.

IV-A. Purification of Oxygen

To reiterate, impurities in oxygen consist of inert gases, e.g., nitrogen, and the noninert gases carbon dioxide, carbon monoxide, methane, and trace quantities of higher molecular weight hydrocarbons. The inert gases are detrimental only in that they will concentrate in the fuel cell and must be purged to maintain acceptable partial pressures of oxygen. The purge stream

carries oxygen, which is waste unless it can be recovered for breathing or other purposes. Our study has not uncovered a practical means to remove the inerts.

The non-inert gases are minor in quantity, and thus are not objectionable from the standpoint of waste through venting. They are quite objectionable in that they are direct or indirect sources of carbon dioxide, which is converted by the fuel cell electrolyte to solid carbonates. Elimination of  $\text{CO}_2$  or  $\text{CO}_2$ -forming impurities is essential for continuous and reliable fuel cell performance. This problem is therefore the most important of those considered in this program.

Analyses of several techniques for removing the  $\text{CO}_2$  family of impurities have shown that the only generally feasible technique consists of conversion of objectionable carbon-bearing gases to carbon dioxide followed by scrubbing out of total  $\text{CO}_2$  content. Success in coping with the problem then hinges on the ease and completeness of oxidation of CO and hydrocarbons, and on the ability of a scrubbing technique to remove  $\text{CO}_2$  to 1 ppm or less. In the broadest sense, these tasks can all be accomplished in theory. In practice, the two most critical problems, CO and  $\text{CO}_2$ , can be coped with without a great amount of difficulty. Methane poses a much greater challenge, one which according to the literature can be met only through the use of quite high temperatures if complete removal is desired.

Three devices, each successively more complex, have been proposed to cope in varying degree with  $\text{CO}_2$  and  $\text{CO}_2$ -forming impurities. Only one of these devices would be employed. Selection of one device in preference to the remaining two should be based on specific knowledge of the nature and quantity of impurities and their effect on fuel cell operation. For example, tests of the effect of methane on fuel cell operation should prove or disprove the need for removal of methane. These arguments are presented in greater detail below in discussions of each of the proposed devices.

IV-A-1. Removal of  $\text{CO}_2$ : The design for the unit as presented in III-A-1-a and in the Appendix should assure  $\text{CO}_2$  removal under all operating conditions. The design is believed to be conservative, however, and developmental studies may well result in worthwhile reductions in volume and weight. As designed, it is our opinion that the  $\text{CO}_2$  scrubber is well worthwhile. Operation of the unit is relatively simple, should be completely reliable, and the weight, volume, and parasitic power penalties are small when balanced against possible cell malfunction.

The chief uncertainty in the operation of this unit lies in the need for the humidifier. The literature is quite positive on this score to the extent that  $\text{CO}_2$  absorption is much more efficient in humid gases than in very

dry gases. Nevertheless, we feel that this question should be explored in the laboratory, as elimination of the humidifier would result in simplification and weight reduction. Further, it is possible that supply oxygen will contain sufficient water to promote  $\text{CO}_2$  absorption. Water in supply oxygen would, however, be very low in concentration in early stages of the mission and increase in concentration as the oxygen storage vessels warmed.

Minor uncertainties arise from the fact that the  $\text{CO}_2$  scrubber is required to operate at elevated pressure rather than the essentially atmospheric pressures used in current devices. We feel that operation will be no different at high pressures, but this point should be checked.

IV-A-2. Removal of  $\text{CO}$ : This unit contains a Hopcalite burner in addition to a humidifier and  $\text{CO}_2$  scrubber. We are confident that the burner will operate effectively as designed. Again, operation under actual mission conditions should be carried out to verify data used in the design, and to determine whether some reduction in size can be tolerated.

IV-A-3. Removal of methane: As mentioned earlier, methane is difficult to oxidize, and the operation of the proposed unit should be thoroughly checked before a design is finalized. The suggested operating conditions will, according to our interpretation of literature data, result in removal of only 60 per cent of the methane. The usefulness of the methane burner is thus debatable, as 10 ppm of methane may be as objectionable as 25 ppm. The operating temperature of the fuel cell,  $450^\circ\text{F}$ , is about the temperature at which methane becomes catalytically oxidizable and it is possible that methane will not be a problem.

It should be pointed out that a Hopcalite burner effectively catalyzes the oxidation of all volatile hydrocarbons except methane at  $500^\circ\text{--}550^\circ\text{F}$ . Other possible hydrocarbon impurities, acetylene for example, are inherently more susceptible than methane to oxidation in the fuel cell. In the event that the oxidizable hydrocarbons are proved to be a problem, their removal could be effected by operation of the catalytic burner at  $500^\circ\text{--}550^\circ\text{F}$ .

In summary, removal of carbon dioxide from oxygen appears to be quite feasible, and the incurred penalties are in our opinion low when balanced against the possibility of cell malfunction. The oxygen purifier is not increased substantially in complexity by addition of a  $\text{CO}$  burner, and the  $\text{CO}\text{--}\text{CO}_2$  package appears to be well worthwhile.

The methane burner will at best incompletely remove methane. Since the methane burner requires substantially elevated temperatures, and combustion of methane is incomplete, the advisability of including a methane burner in the package is debatable. Rough estimates of development costs are presented in Table VIII.

TABLE VIII

ESTIMATED DEVELOPMENT COSTS, OXYGEN PURIFICATION

	<u>CO<sub>2</sub> Absorber</u>	<u>CO and CO<sub>2</sub></u>	<u>CH<sub>4</sub>, CO and CO<sub>2</sub></u>
Laboratory Testing			
Elapsed time, month	6	9	12
Man power, man-years	1	1.5	3
Design, Fabrication, and Testing			
Elapsed time, months	12	12	12
Man power, man-years	1.5	2	4

The estimates reflect our opinions of the relative complexities of various tasks, and are not meant to be rigid. Design, fabrication and testing includes the testing of the operation of a device, and does not include testing as an integral part of the fuel cell module.

IV-B. Purification of Hydrogen

Removal of inert impurities from hydrogen is possible by the membrane diffusion technique. The hydride route should not be seriously considered, since the weight penalty is large, parasitic power consumption appreciable, the technique is operationally complex, supporting data are scarce, and the purification factor is less than satisfactory. The membrane diffusion process is by comparison simple, and offers considerably smaller penalties in weight and power loss. For a given weight of hydrogen, more net power can be realized at the 99.5 per cent purity level with purification than without. However, the weight penalty is approximately the same as that incurred by addition of hydrogen in quantity sufficient to vent the impurities. One concludes, therefore, that redesign of the storage facility and the purge schedule is the preferred technique for handling inert impurities in hydrogen. This conclusion assumes that one is at liberty to redesign these components of the power package, and this is not an assumption the contractor is at liberty to make for the Apollo mission.

We wish to emphasize one feature of the inert impurity problem which is not immediately obvious. Removal processes consist fundamentally of venting or purging procedures, and these must be irreversible and very nearly absolute if they are to compare favorably with direct venting from the fuel cell.

Both the uranium hydride and membrane diffusion processes appear at first glance to be irreversible and absolute, and both would be close to absolute if the output hydrogen could be delivered at very low pressures. With the hydride process, delivery at low pressures means cooling the U-UH<sub>3</sub> bed to about 400°F, and energy requirements become prohibitively high. The membrane diffusion process would be much more conserving of hydrogen at low delivered pressures, but gains would be more than offset by power required to compress the hydrogen to 50 - 100 psia.

We recommend further consideration of the membrane diffusion method only for the Apollo mission, or for other missions with designs sufficiently frozen that the inerts problem cannot be handled by redesign of fuel storage systems and purging procedures. Estimates of time and man power required for development of a membrane diffuser for the Apollo mission are as follows:

#### Securing and Verification of Design Data

Elapsed time: 6 - 12 months  
Man Power: 1.5 - 3 man-years

#### Final Design, Fabrication and Testing

Elapsed time: 12 - 18 months  
Man Power: 2 - 3 man-years

#### Total

Elapsed time: 18 - 30 months  
Man Power: 3.5 - 6 man-years



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## APPENDIX

### UNIT DESIGN CONSIDERATIONS

#### A. Uranium Hydride Process

The basic elements of the uranium hydride process are: (1) a heat exchanger which preheats the impure gas feed by cooling the purified product, (2) a superheater for the impure gas stream, (3) a system of two reactors operating in tandem, and (4) associated controllers, valves and piping. Limiting design cases are presented below:

##### (1) Heat exchanger

Heat transfer	1,500 Btu/hr
Temperature difference	150°F
Transfer area	30 ft <sup>2</sup>
Weight	105 lb.*

##### (2) Superheater

Heat transfer	820 Btu/hr ↔ 240 watts
Weight	30 lb.**

##### (3) Reactors

The reactors have not been thoroughly designed from an energy standpoint because of lack of data. The estimated weight of UH<sub>3</sub> required is 12 lb. Additional heat transfer area between the reactors and casing will probably raise the total to 32 lb.

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\* The high mass required is due to structural problems caused by the 150 psi pressure difference between the hot and cold sides of the exchanger.

\*\* The heater would presumably be a packed ceramic bed imbedded with resistance heaters.

(4) Valves, controllers and piping

These items would be a very significant part of this system due to the tandem reactor operation and difficulties in controlling pressure and temperature of the systems. The weight estimate of 15 lb. would have to be reevaluated after extensive process development on this system.

B. Membrane Diffusion Process

The basic elements of the membrane diffusion process are: (1) a heat economizer, (2) a heater before the diffusion cell, (3) the diffusion cell, and (4) associated instrumentation and piping.

(1) Heat exchanger

Heat transfer	710 Btu/hr
Temperature difference	150°F
Transfer area	15 ft <sup>2</sup>
Weight	50 lb.*

(2) Heater

Heat transfer (by resistance heater)	780 Btu/hr ↔ 235 watts
Weight	30 lb.

(3) Diffuser

Weight	20 lb.
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This piece of equipment is described in the body of this report.

(4) Piping and instrumentation

Weight	5 lb.
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\* Same reason as noted in (A).

The only instrumentation needed is a control of the diffuser temperature and the vent rate. The venting would be on a continuous basis.

### C. Removal of Carbon Dioxide

This system consists of a small heater to warm the oxygen stream to the desired humidification and absorption temperature (50°F), a bed of moist silica gel to humidify the gas, and LiOH absorber to scrub the CO<sub>2</sub> from the gas stream. The system is sized to remove as much as 120 ppm of CO<sub>2</sub> from the gas at 8 per cent relative humidity. The specifications of the units are:

#### (1) Heater

Temperature increase	185°F
Heat transfer	135 Btu/hr $\longleftrightarrow$ 40 watts
Weight	5 lb.

#### (2) Humidifier

Material	Silica gel (0.35 lb. H <sub>2</sub> O/lb activated silica gel)
Residence time	1 min. (150 psia, 60°F)
Mass of silica gel	4 lb.
Volume	0.083 ft <sup>3</sup>
Size (cylindrical)	12 in. x 4 in. (O.D.)
Weight of unit	6 lb.

#### (3) Absorber

Material	granular LiOH
Residence time required	0.2 min.*

\* This requires only 0.3 lb. LiOH, however the system was oversized by a factor of 3.

Weight used	1.0 lb. LiOH
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Total weight	3 lb.
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(4) Controller and piping

Weight	3 lb.
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D. Low Temperature Oxidation

All equipment here is the same as that shown in (C) with the exception of a slightly larger heater load and the use of the Hopcalite bed to perform the oxidation. The design was based on 90 per cent conversion of CO to CO<sub>2</sub> at maximum flow and the kinetic data of Mulay.<sup>10/</sup>

Additional heat	2 watts
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Weight of catalyst bed	5 lb.
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Residence time	1 min.
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E. High Temperature Oxidation

This system has the same humidification and absorption equipment as (D) but the initial part of the system is completely different due to the higher temperature of the reactor. This part consists of three pieces: (1) the exchanger, (2) a heavy duty heater, and (3) the burner.

(1) Heat exchanger

Heat transferred	358 Btu/hr
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Temperature difference	200°F
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Heat transfer area	6 ft <sup>2</sup>
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Weight	25 lb.
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(2) Heater

This unit supplies about the same amount of heat as the unit in (C-1). However the heat must be supplied at a much higher temperature and might therefore cause some design changes.

Heat supplied 20 watts

Weight 10 lb.

(3) Burner

This unit is actually smaller than that in (D). The catalyst bed weighs only 3 lb. and residence time is about one-half that in (D). The assumed weight is 5 lb.